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PAPER VII

Radiochemical Analysis of the Bikini Ashes

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I. INTRODUCTION

Radioactive ashes produced by the hydrogen bomb test at Bikini Atoll on March 1, 1954 happened to fall on a Japanese fishing boat, the No. 5 Fukuryu Maru, which was then approximately 90 miles from Bikini.* Twenty-three crew members on board suffered from radiation sickness by this accident, which was brought to light after the boat returned to her home port of Yaizu on March 14, 1954. The present work was undertaken because it was then urgently necessitated to disclose the nature of the radioactive ashes in order to obtain some knowledges which might be valuable for the medical treatment for the afflicted fishermen.**

Two methods were employed to separate and assign the elements in the ashes. The one is the combined method of the usual analysis employing carriers and separation with the ion exchanger. The other is the method using only the ion exchanger. After the chemical separation the assignment of nuclides was carried out by observing the radioactive properties of the species by means of the end-window G-M counters and the scintillation counter. A brief account of the study on radioactivities in the rain at Kyoto on May 16 is also given in the fourth chapter of this paper.

II. PHYSICAL MEASUREMENTS

1. Apparatuses

The physical assignment of the nuclides separated chemically was made by the determination of β - and γ -ray energies from the samples by the absorption method using end-window G-M counters and a scintillation counter. The end-window G-M counter used in this work had the cathode with an inside diameter of 20 mm and a

* This position was outside the demarcation of the "danger zone" from which the United States had long since warned all shippings.

** On this occasion the authors wish to express their deep appreciation to Prof. T. Shiokawa of Shizuoka University, Prof. K. Kimura of Tokyo University, Dr. T. Maekawa, Chief of Sanitation Division of Shizuoka Prefecture, and staffs of Yaizu Municipal Office for their kind aids in collecting the radioactive ashes from the No. 5 Fukuryu Maru.

mica window of 3 mg/cm^2 or 1.5 mg/cm^2 in thickness*. The scintillation counter** used for γ -ray absorption had terphenyl in xylene solution as a phosphor. Pulses from the counters were measured with a scale-of-100 scaler with a mechanical recorder. Some precautions were taken to keep the standard error in the counting rate for each measurement below about 3 per cent.

The G-M tube and shelf assembly and the arrangement of γ -ray absorption with the scintillation counter are shown in Figs. 1 and 2, respectively.

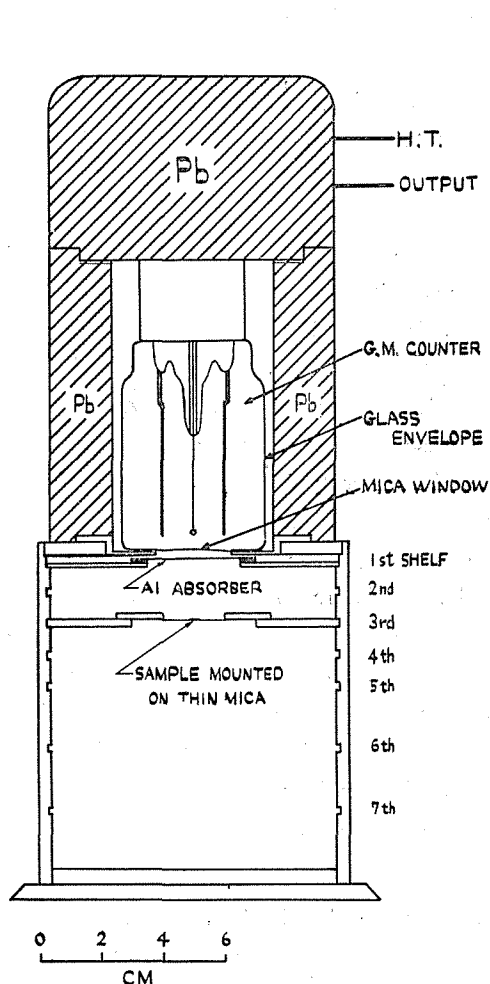


Fig. 1 G-M counter and shelf assembly for β -ray absorption.

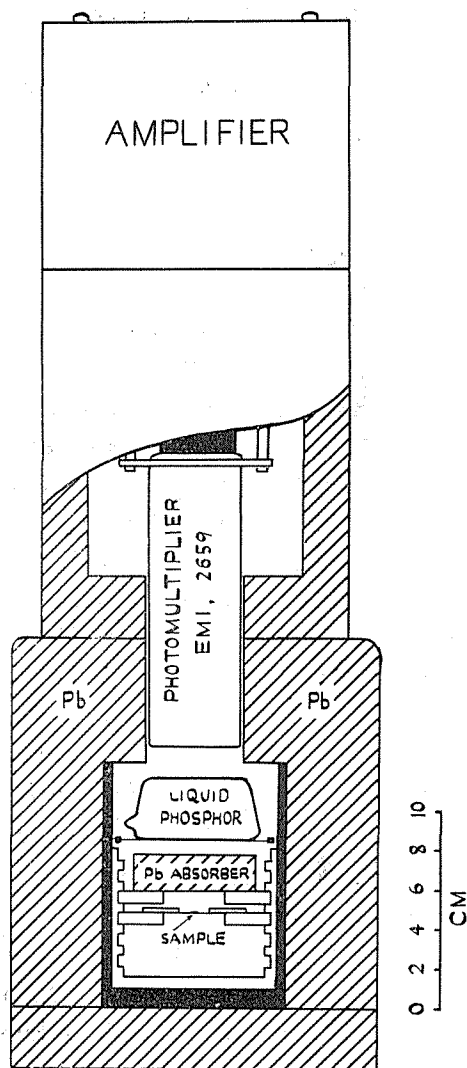


Fig. 2 Arrangement of γ -ray absorption by the scintillation counter.

* Manufactured by Kobe Kogyo Co. Ltd., Kobe, Japan : GM-131 (3 mg/cm^2) and GM-132 (1.5 mg/cm^2).

** Manufactured by EKCO Electronics Ltd., Southend-on-Sea, Essex, England: Type N502

2. Determination of β -ray energies.

The determination of β -ray energies was carried out by observing the half-thickness for aluminum and by estimating the total range in aluminum by the application of the Feather method of analysis. The empirical curve for the relation between half-thickness and energy, used in the present work, was obtained in such a geometry that the samples were placed on the 2nd shelf and the absorbers on the 1st shelf, by using C^{14} , P^{32} , Co^{60} , Sr^{89} and I^{131} which were available in our laboratory. This geometry of measurement was decided by the reason that the half-thickness method is applicable for the samples with comparatively low intensities. For some samples we applied the Feather analysis, in such cases the samples were placed on the 3rd shelf while the absorbers on the 1st shelf. The Feather analyzer strip necessary for this method was prepared by observing the β -ray absorption of Y^{91} as a reference species, which was separated from the fission products and purified by the ion exchange technique, since we were not able to use RaE, generally used for this purpose. This is illustrated in Fig. 3 and Table 1.

To avoid the variation of the β -ray spectrum due to the backscattering of β -rays from the sample holder as possible, the sample was placed on a thin mica

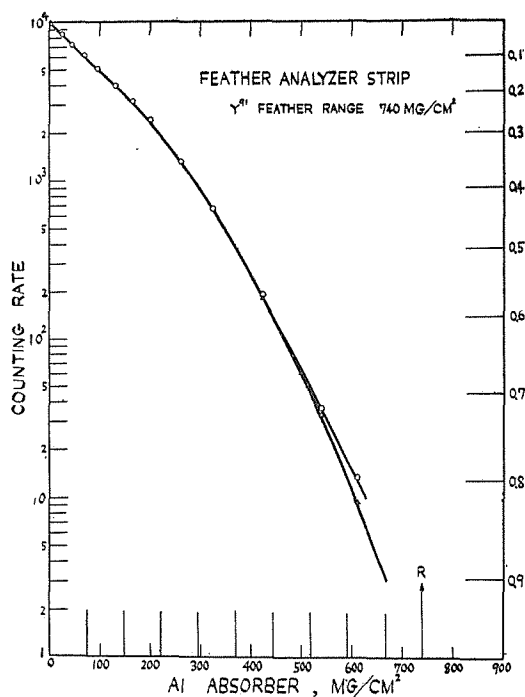


Fig. 3 Aluminum absorption curve of the β radiations of Y^{91} . \circ , values in the column A ; \triangle , values in the column B in Table 1. Strip on right side represents the Feather analyzer for the standard counting arrangement. Source is on the 3rd shelf with absorbers on the 1st shelf.

Table 1. Relative counting rates through absorbers for the Feather analysis of standard absorption curve of Y^{91} .

mg/cm ²	A	B	mg/cm ²	A	B
0	10000	9996	164.7	3192	3188
13.4	8889	8885	199.8	2409	2405
25.04	8423	8419	259.2	1326	1322
46.7	7091	7087	324.0	679.5	674.5
70.2	6186	6182	423.9	197.4	193.4
95.2	5115	5111	540.0	37.3	34.3
129.6	4065	4061	610.2	13.7	9.7

A : counting rate (natural background is subtracted)

B : counting rate (the background of the scattered radiation is subtracted)

sheet less than 5 mg/cm² in thickness as shown in Fig. 1. The sample in solution was dropped on the mica sheet and evaporated to a thin layer, taking the self-absorption and self-scattering into consideration. When the sample contained a large amount of organic compounds, it had to be decomposed with $HClO_4$ to reduce the volume to be measured. In addition to this, since the absorption in the counter window is marked for low energy β -rays, a G-M counter with a mica window of 1.5 mg/cm² was used for the β -rays less than 0.3 Mev. However, in spite of these cautions the reliable absorption curve for the β -ray energies below 0.15 Mev could not be obtained. The β -ray absorption curves shown in the figures in the following chapters were obtained by these procedures.

The physical assignment of nuclides by the determination of β -ray energies may be quite troublesome when the unknown sample to be measured contains some other radioactive species. When the contamination was not extreme and its β -ray energies were considerably higher than those of the main nuclide, the counts due to the contamination were subtracted from the original curve as the background. For instance, the absorption curve of Zr^{95} containing the rare earth of about 50 per cent is shown in Fig. 15, while in Fig. 22 that for the purified sample is shown, which contains still the rare earths of about 2 per cent.

3. Determination of γ -ray energies.

Often we could not ascertain what kind of nuclide existed in the sample even by the measurement of β -ray absorption, especially in the case that the sample contained the daughter products. In such cases it was often helpful to measure the γ -ray energies.

The determination of γ -ray energies was performed by the absorption in lead, using a scintillation counter with terphenyl in xylene solution, 5 cm in diameter and 2.5 cm in depth, as a phosphor. The arrangement of the apparatus is shown in Fig. 2. By this procedure the curves in Figs. 21, 23 and 25 were obtained. The absorption curve for γ -rays from La^{140} , daughter of Ba^{140} , was obtained by observing Ba

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sample after the radioactive equilibrium of both nuclides had been attained.

4. Measurement of decay.

The decay of the Bikini Ashes and several species separated chemically from the ashes was measured by the end-window G-M counter. The special assembly for this purpose was constructed as shown in Fig. 4. The sample, placed on an alumi-

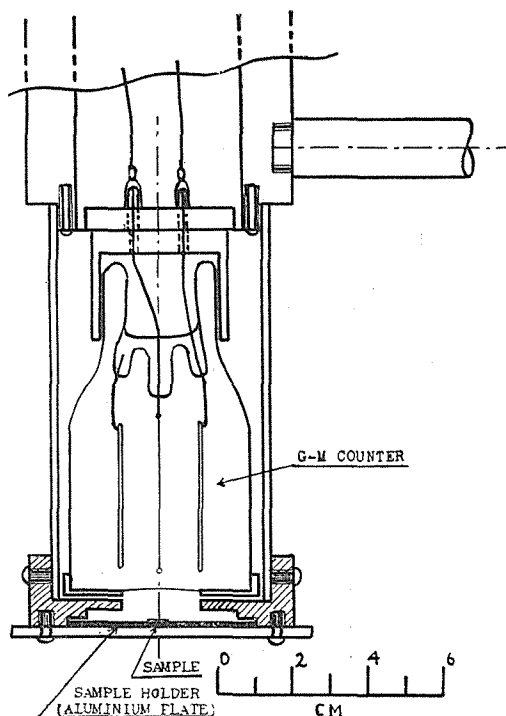


Fig. 4 Counting arrangement for decay measurement.

num plate, 1 mm in thickness and 5 cm×7 cm in size, was inserted in the guide to a fixed position. By such device we could maintain the counting geometry constant all over the period of the measurement. The slight day-by-day variation of the sensitivity of the counter was calibrated by employing Co^{60} and U_3O_8 reference sources. The probable error in each counting rate was kept less than 2 per cent.

The decay of two samples of the Bikini Ashes, one was white and the other was dark grey, has been measured since May 4, i.e. the 64th day after the explosion; the decay curves obtained are shown in Fig. 5. Following empirical expressions for the activity were derived from our measurements for the interval of 81 days :

$$A_t = A_0 t^{-1.17 \pm 0.02} \quad \text{for the white ashes,}$$

$$\text{and } A_t = A_0 t^{-1.24 \pm 0.05} \quad \text{for the dark grey ashes,}$$

where A_t is the activity at time t after the explosion and A_0 is a constant.

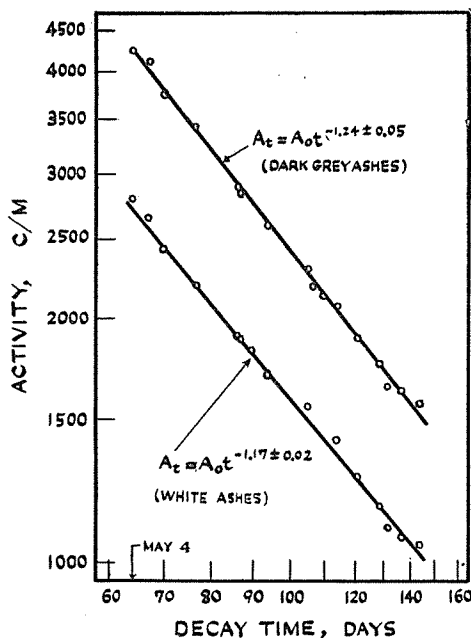


Fig. 5 Decay curves of the Bikini Ashes.

It may be interesting to compare our expressions with the similar one given by other workers for fission products of U^{235} by the irradiation of slow neutrons.^{1,2)}

The decay curves obtained for Y^{91} , Ba^{140} - La^{140} , and U^{237} are shown in Figs. 13, 26 and 29, respectively. Our half-life of U^{237} is in good agreement with that given by other workers,³⁾ however, that the half-life of Ba^{140} from our data in Fig. 26 is slightly larger than the value given by other workers,⁴⁾ may be explained by the presence of some impurities.

III. CHEMICAL ANALYSIS

1. Preparation of sample

The sample gathered from the deck of the No. 5 Fukuryu Maru was dark grey dust which contained wooden pieces and iron rust. The radioactivity resulted from the fission products attached to small fragments of coral reef of Bikini Island. The decay of its activities was measured as shown in Fig. 5, and the β -ray absorption curve observed is shown in Fig. 6. The sample was moistened with dil. NaOH and ashed in a porcelain crucible. The ashes were fused with Na_2CO_3 - K_2CO_3 , and the fused mass was treated with hot water and digested on a water bath for one hour. After the complete elimination of the phosphates had been ascertained with ammonium molybdate, the sample was taken up with conc. HCl. Carrier solutions*

* Nitrates of Se, Y, Zr, Nb, Sr, Ag, In, Rh, Sb, Ba, La, Ce, Pr, Nd, Er and Yb. Chlorides of Pd, Cd and Sn. 1/100 m. mole of each element was added.

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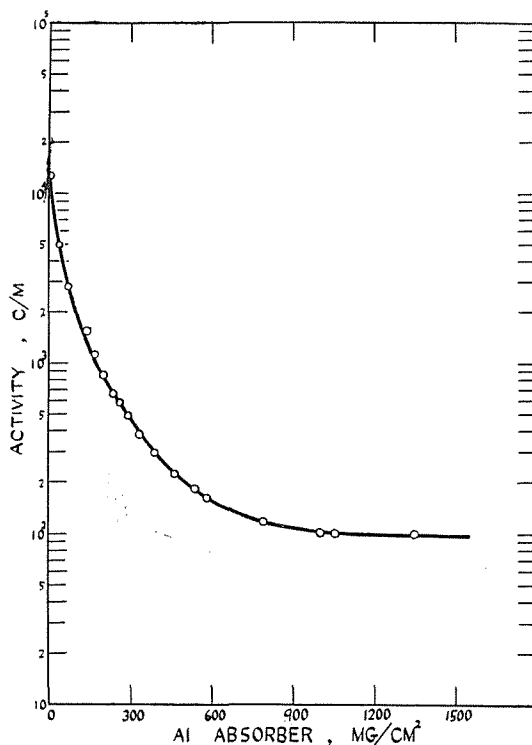


Fig. 6 Aluminum absorption curve of the Bikini Ashes.
(Dark grey ashes, March 25, 1954)

were added and evaporated to dryness on a water bath.

In our analysis the special precaution was taken to remove the silicates completely in order to avoid the following difficulties : In the separation of elements using the coarse grained ion-exchange resin, the colloidal silica, carrying radioactive species, would pass through the exchanger so that radionuclides would be eluted irregularly, accompanying undesired contaminations. When the fine grained ion exchange resin is used, the colloidal silica would be left on the upper part of exchanger bed, adsorbing considerable portion of radionuclides.

2. Analysis by the method of the conventional grouping.

The group separation was carried out by the conventional H₂S method,⁹ as illustrated in the flow sheet shown in Fig. 7. For further separation of each group, the ion exchange technique was also applied.

a) The first group : The residue insoluble in 4N HCl was taken up to dil. NH₄OH, then NaOCl solution was added to the filtrate, followed by evaporation to dryness. This sample was identified as I¹³¹ by measuring the β -ray absorption as shown in Fig. 8.

b) The second group : The acidity of the solution was adjusted to 0.3N, and the elements of the second group were precipitated as sulfides.

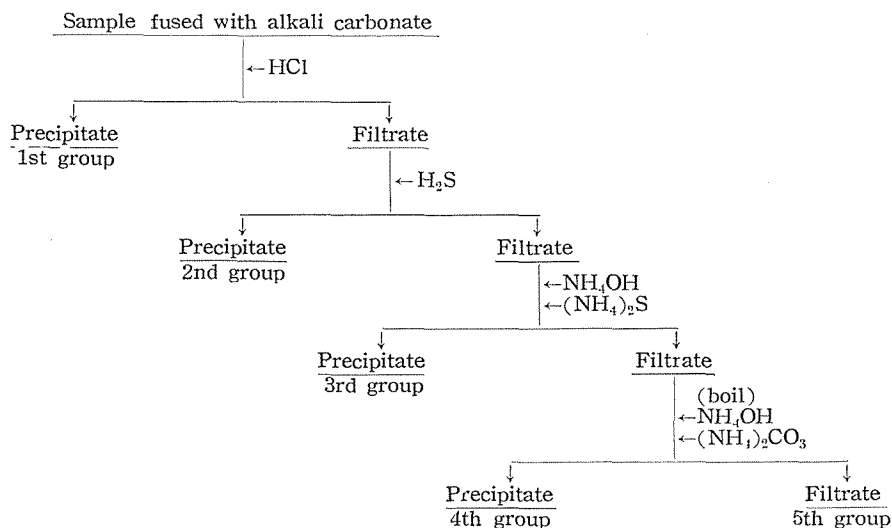


Fig. 7 Flow sheet showing analytical procedure for group separation of the radioactive dust.

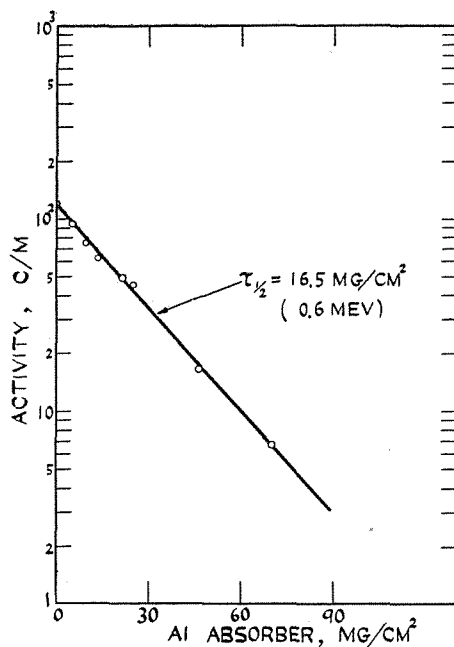


Fig. 8 Aluminum absorption curve of I^{131} .

The sulfides were dissolved in HNO_3 . A saturated alcoholic solution of p-dimethyl aminobenzylidene rhodanin was added drop by drop under vigorous stirring to the solution at pH 1.0. The precipitate of Pt-group separated by this procedure had no activity.

Se and Te were reduced to the metallic state with ferrous sulfate and hydrazine

hydrochloride solution, respectively.⁶⁾ The radioactivity of Se obtained here was too weak to measure its energy. By observing the β -ray absorption of precipitated Te, Te^{129} was assigned as shown in Fig.9.

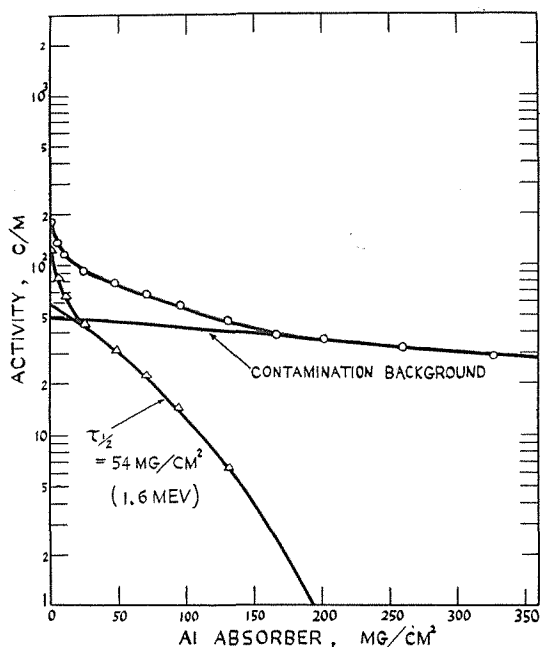


Fig. 9 Aluminum absorption curve of Te^{129m} - Te^{129} .
○, original points. △, contamination background subtracted.

The organic matter in the filtrate was decomposed by treatment with conc. HNO_3 . The residue was taken up with HCl and the acidity was adjusted to 0.3 N. The solution was saturated with H_2S ; no activity was found in the precipitate of the Cu-group thus obtained.

c) The third group : Using the filtrate from the second group elements, the third group was precipitated as sulfide. The HCl solution of the third group at a pH of 1.0 was adsorbed on the resin bed ($120\text{ cm} \times 1.13\text{ cm}^2$) of the H-form Amberlite IR-120 (40~60 mesh),* and the width of the adsorption band was about 5 mm. After washing with water, radioactive species were eluted out using the following solutions successively at a flow rate of 1.0 ml/min/cm².

* Ion exchange resin beds used in the present work were prepared as follows: Amberlite IR-120 resin was crushed and sieved to the suitable size for each elution. After the resin was warmed with 4N HCl on a water bath to convert it into H-form and to swell, it was packed in a pyrex glass column, bottom of which was closed by glass wool, as shown in Fig. 10. The resin bed was washed with 1N HCl , and then with water to remove the excess HCl completely.

Order of elution	Elutrient	Eluted species
1	0.2N HNO ₃	Radiocolloids
2	0.5% Oxalic acid (pH 1.7)	Fe and Radiocolloids
3	5% Amm.-citrate (pH 3.85)	Rare earths

The each 10 ml fraction of effluent was received in crucibles and evaporated to dryness with addition of H₂O₂ and HClO₄ to decompose the organic compound.

The eluate of rare earths dried was again dissolved in HCl and pH was adjusted to 1. Then, it was adsorbed on the ion exchanger bed (45 cm × 0.50 cm²) of the H-form Amberlite IR-120 (150~250 mesh), as shown in Fig. 10.

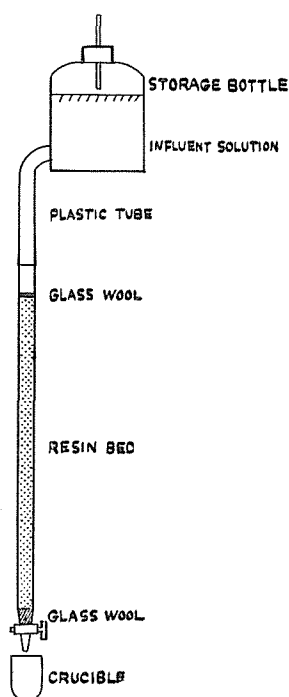


Fig. 10 Resin column : the flow rate is adjusted by varying the height of the storage bottle.

The rare earths were eluted with 5 % citrate solution (pH 3.2) at a flow rate of 0.08~0.1ml/min/cm²). The effluent was collected in porcelain crucibles at an interval of 30 minutes. The "cut" in each crucible was dried and activities were measured. The elution curve of active rare earths is shown in Fig. 11.

A few "cuts" corresponding to each peak in the elution curve were collected respectively, and treated with HClO₄ and evaporated to dryness. The β -ray absorption curves were obtained for these dried residues as shown in Figs. 12 and 14. Fig. 13 shows the decay observed for Y⁹¹. By these physical measurements Y⁹¹, Pr¹⁴⁴ and Ce¹⁴⁴ were assigned.

The eluted radiocolloids were treated with H₂O₂-HClO₄, and the excess of

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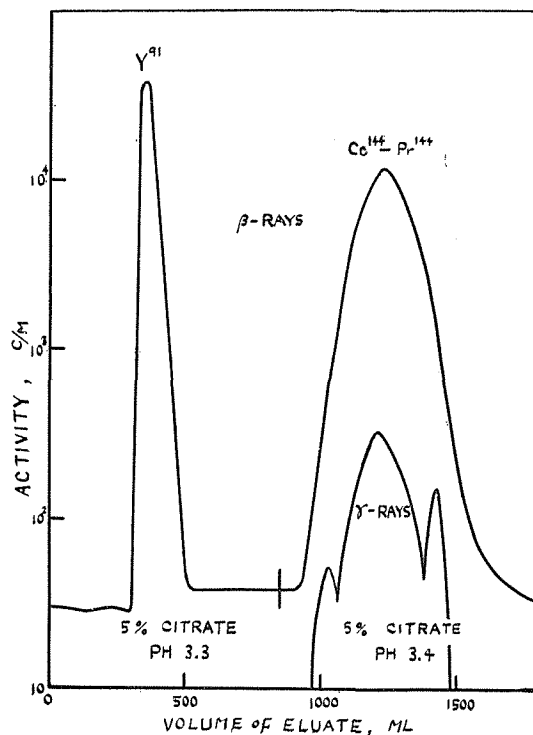


Fig. 11 Separation of the rare earths in the Bikini Ashes by eluting through 150~250 mesh Amberlite IR-120 ; bed dimension 45 cm \times 0.50 cm² ; flow rate 0.1 ml/min/cm². For γ -ray curve the noise of the scintillation counter is subtracted.

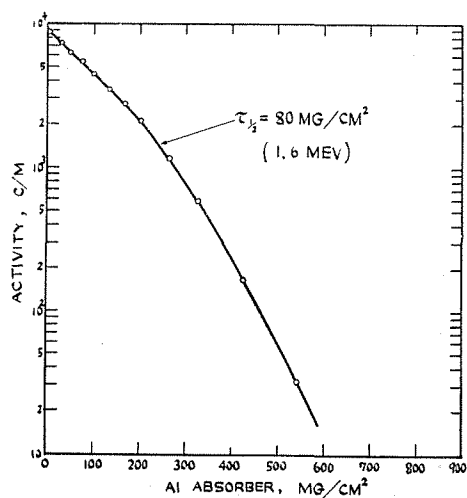


Fig. 12 Aluminum absorption curve of Y^{91} .

$HClO_4$ was expelled by evaporating with several portions of HCl . Ammonium hydroxide was added and the precipitate was fused with alkali carbonate for three

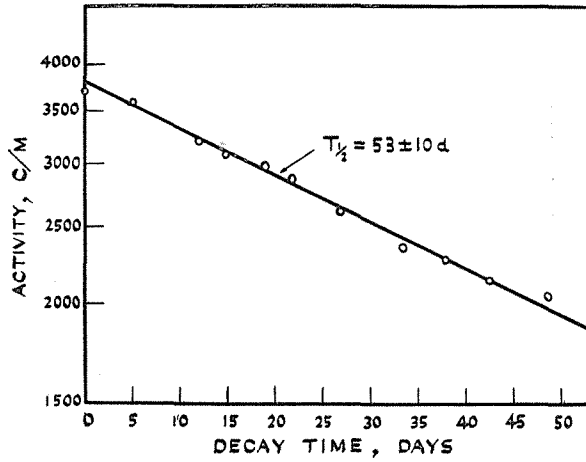


Fig. 13 Decay curve of Y^{91} .

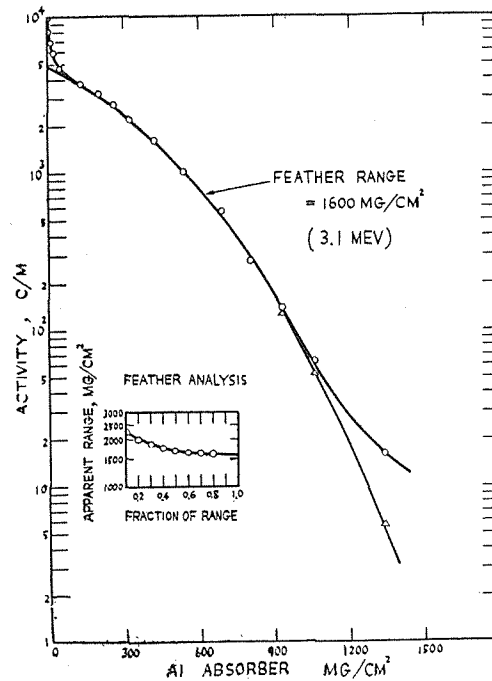


Fig. 14 Aluminum absorption curve and Feather analysis of Ce^{144} - Pr^{144} equilibrium mixture. \circ , original points. \triangle , γ background subtracted. Feather analysis shows β radiations (3.1 Mev) of Pr^{144} .

hours. The fused mass was leached with 1N NaOH on a water bath, and the supernatant was acidified with a little excess of HCl to precipitate hydrated Nb_2O_5 , which was washed with dil. HCl several times and dried. Nb could not be identified in this sample, because its activities were too weak to measure. However, Zr was

isolated from the residue with tannic acid according to the Schöller method.⁸⁾ The β -ray absorption curve of Zr^{95} is shown in Fig. 15.

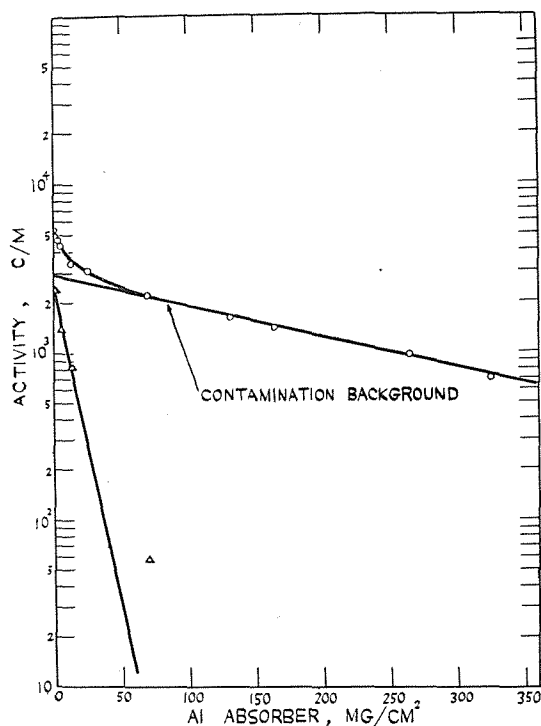


Fig. 15. Aluminum absorption curve of Zr^{95} . \circ , original points. \triangle , contamination background subtracted.

d) The fourth group : Using the filtrate from the third group, the elements of the fourth group were precipitated as carbonates and contaminations were removed by coprecipitation with FeS . The carbonate of the fourth group was dissolved in HCl and evaporated to dryness. Then, the residue was taken up with $0.3\text{N } \text{NH}_4\text{OH}$.

Alkali earths were adsorbed on the ion exchanger bed ($30 \text{ cm} \times 0.28 \text{ cm}^2$) containing NH_4 -form⁺ Amberlite IR-120 (100~150 mesh), the width of adsorption band being about 5 mm. The flow rate was 0.1 ml/min/cm^2 . Alkali earths were separated with the following elutriants.

Order of elution	Elutrient	Eluted Species
1	1.3 N Amm.-acetate	Ca
2	2 N Amm.-acetate	Sr
3	ETA 5 ml + 2 N Amm.-acetate 5 ml + Ammonia (1 : 1) 2 ml	Ba

The elution curve of alkali earths is shown in Fig. 16. The activity of Ba was too weak to assign the nuclide.

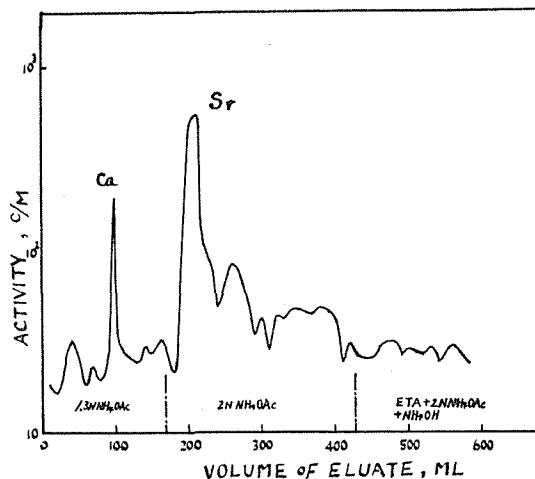


Fig. 16 Separation of the alkali earths in the Bikini Ashes by complex elution : Amberlite IR-120 (100~150 mesh) ; bed dimension 30 cm \times 0.28 cm² ; flow rate 0.1ml/min/cm² ; elutrient, 1.3 N amm.-acetate for Ca, 2 N amm.-acetate for Sr.

The β -ray absorption curves of Ca⁴⁵ and Sr⁸⁹ are shown in Figs. 17 and 18, respectively.

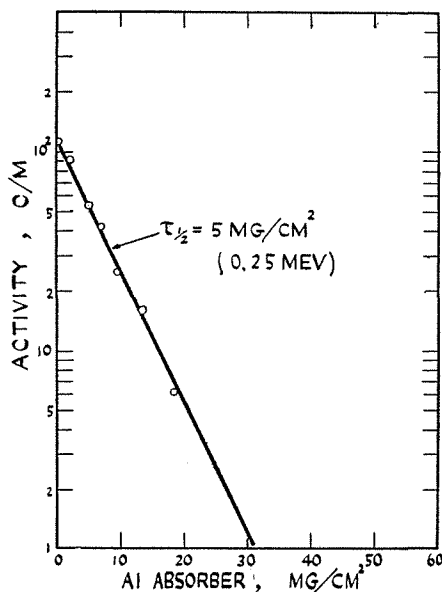


Fig. 17 Aluminum absorption curve of Ca⁴⁵.

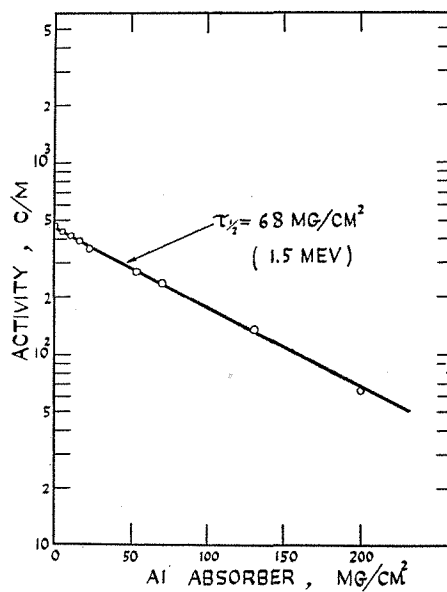


Fig. 18 Aluminum absorption curve of Sr⁸⁹.

e) The fifth group : Using the filtrate from the fourth group, most part of Na and K were successively separated with methyl acetate and tartrate, respectively. No activity was observed in Na and K fractions, while the residual part had so weak

activity that Rb and Cs could not be identified.

3. Analysis by the method of the ion exchange grouping

The group separation was carried out by the use of ion exchange technique (cf. foot-note in page 47), since this technique was found to be very effective for the analysis of radioactive rain-water, as mentioned in the following chapter.

a) Group separation of radioactive species : The radioactive dust was treated with $\text{H}_2\text{O}_2\text{-HClO}_4$ to decompose the organic matter, and the solution was evaporated. The residue was taken up with several portions of conc. HCl, and the acidity of the chloride solution was adjusted to pH 1, which was then adsorbed on the ion exchanger bed ($20\text{ cm} \times 0.28\text{ cm}^2$) of Amberlite IR-120 (100~150 mesh). The group separation was carried out by passing the following elutriants through the column successively with a flow rate of 0.2 ml/min/cm^2 .

Order of elution	Elutiant	Eluted Species
1	0.1N HCl	Anions
2	0.5% Oxalic acid	Radiocolloids
3	5% Amm.-citrate (pH 3.85)	Rare earths
4	5% Amm.-citrate (pH 5.90)	Alkali earths

Each "cut" (10 ml) of the effluent was collected in porcelain crucibles and evaporated to dryness. The β -ray activity of each crucible was measured and the result

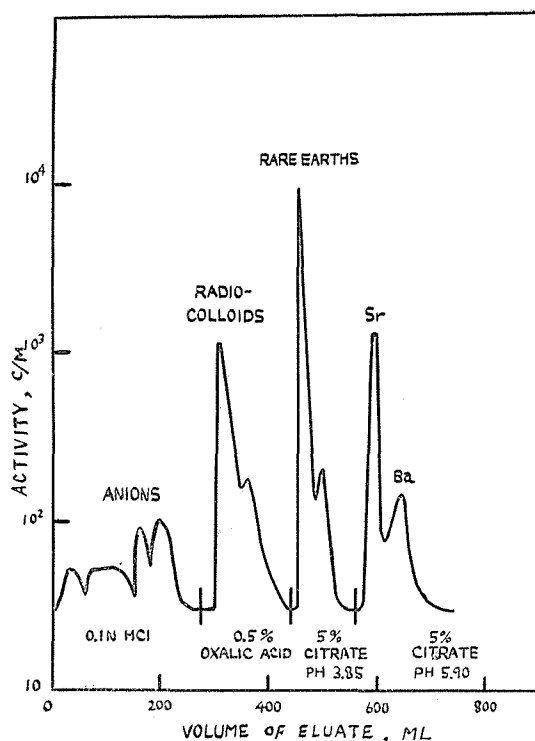


Fig. 19

Group separation of the radioactive elements in the Bikini Ashes by an ion exchanger : Amberlite IR-120 (100~150 mesh); bed dimension $20\text{ cm} \times 0.28\text{ cm}^2$; flow rate 0.2 ml/min/cm^2 .

is plotted in Fig. 19.

b) Assignment of Ru and Rh : The eluate of the anion group was evaporated on a mica plate. From observation of the β -ray and γ -ray absorption for this sample, Ru^{103} , Rh^{103m} , Ru^{106} and Rh^{106} were assigned, as shown in Figs. 20 and 21.

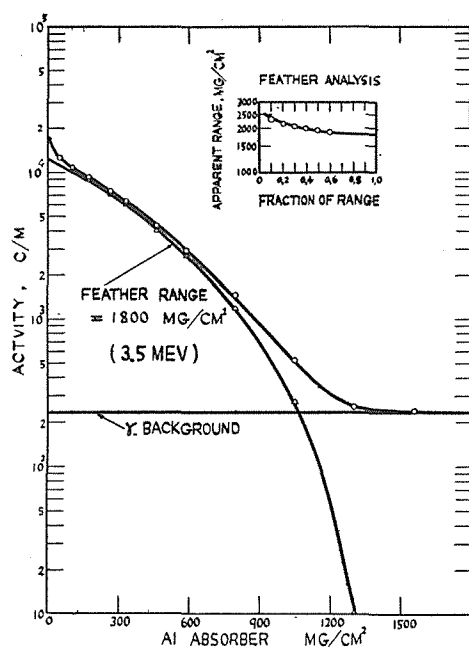


Fig. 20 Aluminum absorption curve of Ru^{103} - Rh^{103m} , Ru^{106} - Rh^{106} equilibrium mixture which was separated chemically as Ruthenium. Feather analysis shows β radiations (3.5 Mev) of Rh^{106} .

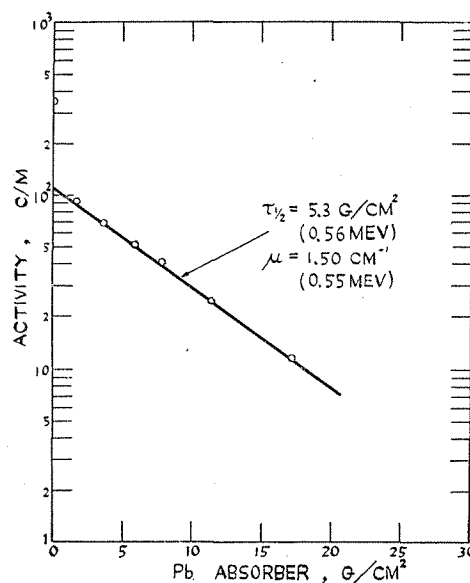


Fig. 21 Lead absorption curve of Ru^{103} - Rh^{103m} equilibrium mixture. Curve shows γ -rays of Ru^{103} .

c) Purification of Nb and Zr : The oxalate solution of Nb and Zr eluted was evaporated with HClO_4 to dryness to decompose organic materials completely. The residue was moistened and dried several times with HCl to convert Nb and Zr into chlorides, and taken up in 10 ml of 6N HCl . From this solution Zr-Nb was extracted as cupferrate with CHCl_3 ⁹⁾, and then returned to aqueous phase after washing with cupferron and HCl . As shown in Fig. 22, this sample was less contaminated than that obtained by tannin method (cf. Fig. 15). Gamma-ray absorption curve for this sample is also given in Fig. 23.

d) Assignment of Ba : After the decomposition of organic materials in the effluent, Ba was precipitated as carbonate accompanying other alkali earth elements. From the alkali earth mixture, Ba was isolated by treatment with HCl -ether (1:1) at low temperature¹⁰⁾. Figs. 24 and 25 show the β - and γ -ray absorption curves for this sample, respectively, thus Ba^{140} and La^{140} were assigned. The decay curve of Ba^{140} - La^{140} is shown in Fig. 26.

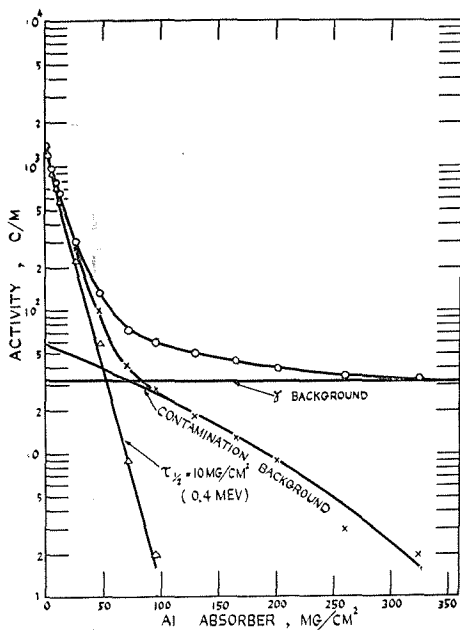


Fig. 22 Aluminum absorption curve of Zr^{95} .
 \circ , original points. \times , γ background subtracted. \triangle , γ and contamination background subtracted.

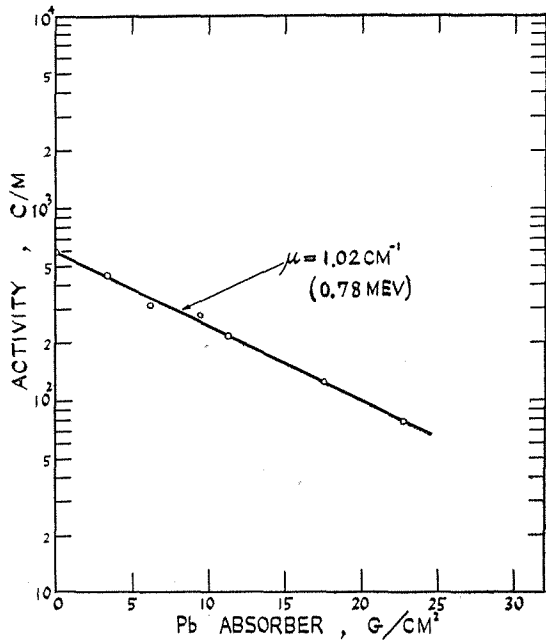


Fig. 23 Lead absorption curve of Zr^{95} - Nb^{95} .

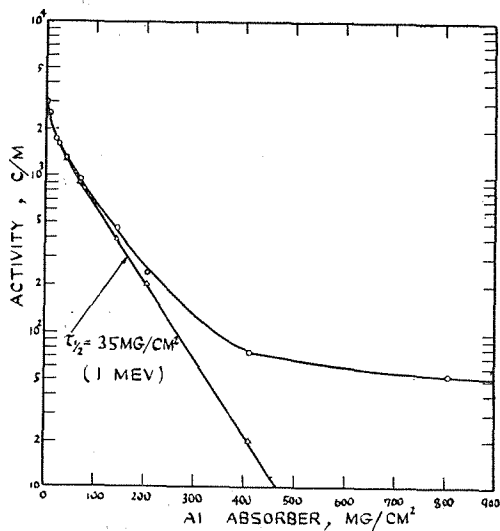


Fig. 24 Aluminum absorption curve of Ba^{140} measured immediately after separation.

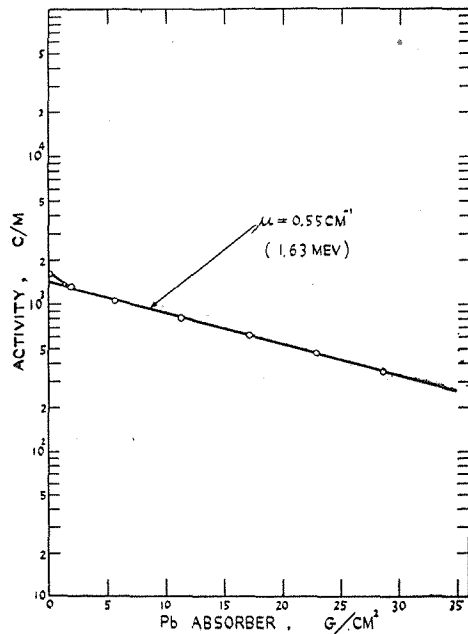


Fig. 25 Lead absorption curve of Ba^{140} - La^{140} equilibrium mixture. Curve shows γ -rays of La^{140} .

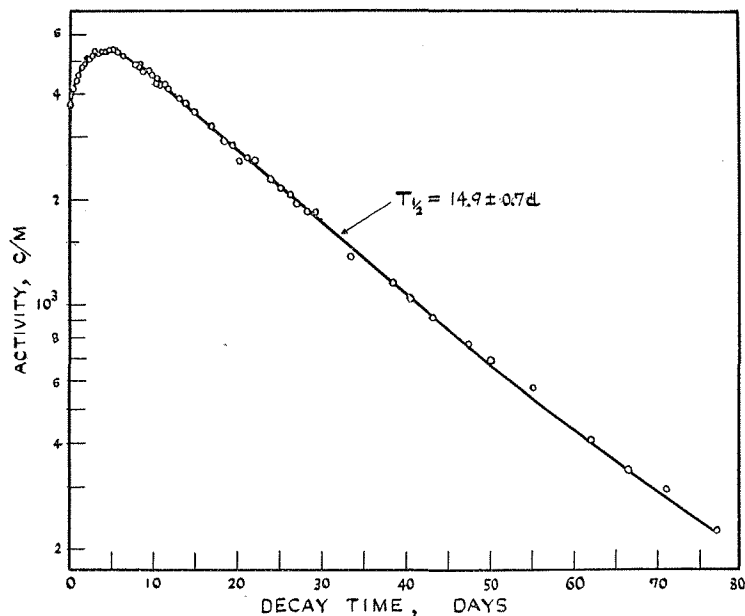


Fig. 26 Decay curve of Ba¹⁴⁰-La¹⁴⁰.

4. Separation of U²³⁷ by paper chromatography

One drop of the concentrated sample solution (cf. 3-a) was spotted on a paper strip (Toyo Filter Paper, No. 50), and developed by HCl-butanol solution (1 : 3) for 16 hours at room temperature. The β -ray activity of the strip was measured by an end-window G-M counter through a slit of 5 mm width slotted in a lead sheet of 2 mm thickness. The result is plotted in Fig. 27. An intense peak of activity was found at the position corresponding to $R_f=0.54$. The β -ray absorption curve and decay curve of this activity are shown in Figs. 28 and 29, respectively.

It may be interesting to note that from these results the activity of this peak seems to be attributed to U²³⁷, which emits β -rays of 0.24 Mev and has the half-life of 6.63 days³⁾.

IV. ANALYSIS OF RAIN-WATER AT KYOTO CITY ON MAY 16, 1954.

The rain-water at Kyoto City on May 16, 1954 was found to have intense radio-activities of about 1 μ C/l. The radioactive elements in the rain-water of about 250 ml* were separated by the similar procedure as described in III, 3-a, using ion exchanger. The result is shown in Fig. 30, and the presence of Sr⁸⁹, Ba¹⁴⁰, Nb⁹⁵ and Zr⁹⁵ was ascertained by β -ray absorption curves, as shown in Figs. 31, 32 and 33.

Of these nuclides we were able to determine the absolute activity for only Zr⁹⁵ to

* The rain-water was collected by Prof. T. Shidei, Department of Applied Physics, Kyoto University.

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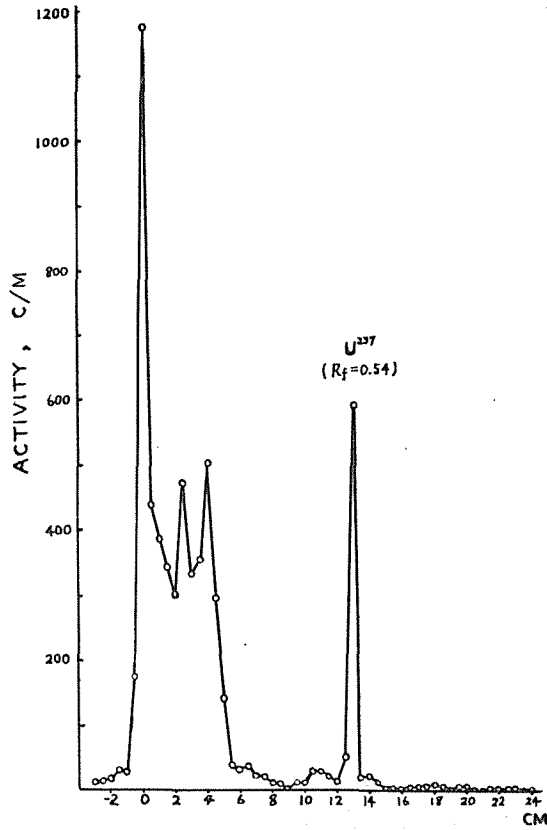


Fig. 27 Elution curve of paper chromatograph.

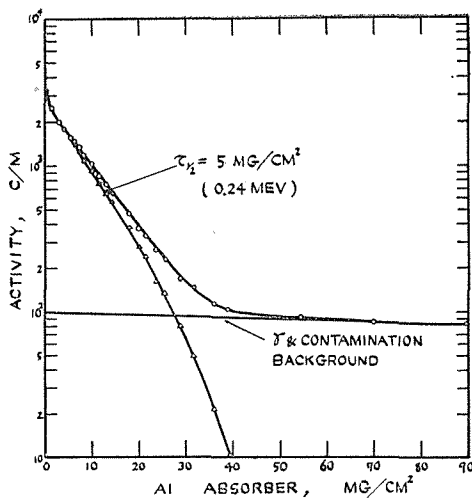


Fig. 28 Aluminum absorption curve of U^{237} . \circ , original points. Δ , γ background subtracted.

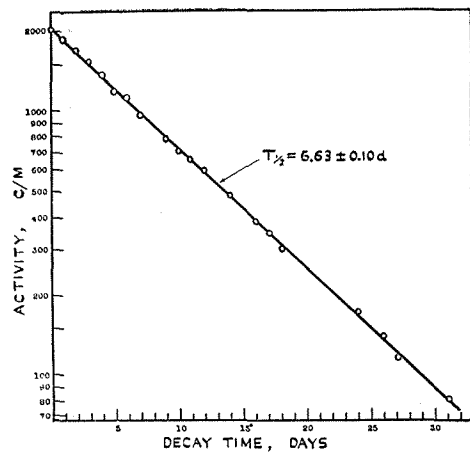


Fig. 29 Decay curve of U^{237} .

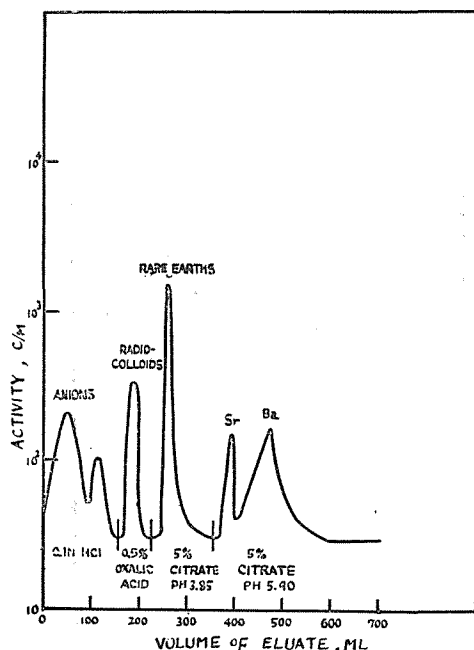


Fig. 30 Group separation of the radioactive elements in the rain-water (Kyoto City, May 16, 1954) by an ion exchanger: Amberlite IR-120 (100~150 mesh); bed dimension 20 cm \times 0.28 cm²; flow rate 0.2 ml/min/cm².

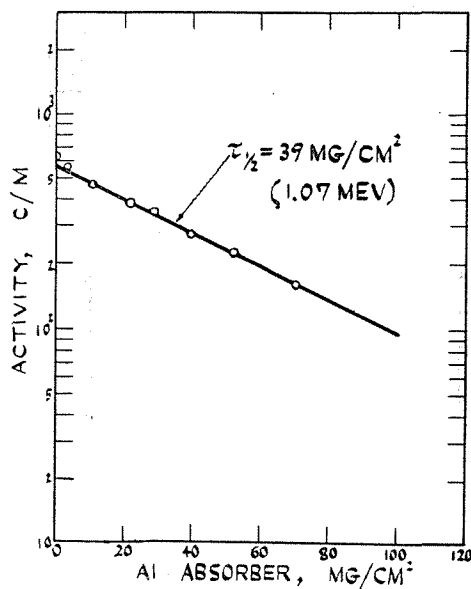


Fig. 32 Aluminum absorption curve of Ba¹⁴⁰ in the rain-water.

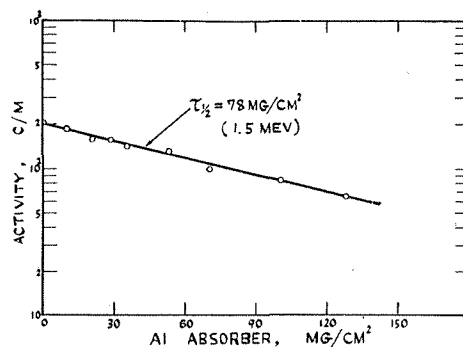


Fig. 31 Aluminum absorption curve of Sr⁸⁹ in the rain-water.

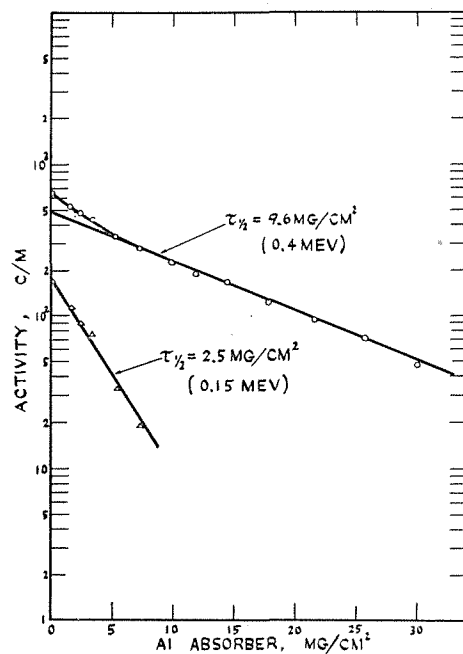


Fig. 33 Aluminum absorption curve of Zr⁹⁵ and Nb⁹⁵ in the rain-water. \circ , Zr⁹⁵. \triangle , Nb⁹⁵.

be $5.7 \times 10^{-2} \mu\text{C} \pm 20\%$ per liter, taking many factors concerning the absolute β counting into consideration.

V. SUMMARY

From the Bikini Ashes following nuclides were assigned by the radiochemical procedure; Ca^{45} , Sr^{89} , Y^{91} , Zr^{95} , Nb^{95} , Ru^{103} , $\text{Rh}^{103\text{m}}$, Ru^{106} , Rh^{106} , Te^{129} , I^{131} , Ba^{140} , La^{140} , Ce^{144} , Pr^{144} and U^{237} . The ion exchange method was very effective for the analysis of rain-water, from which the presence of Sr^{89} , Zr^{95} , Nb^{95} and Ba^{140} was ascertained, while the presence of rare earths was also found but their assignments could not be carried out.

Comparing the elution curves for the Bikini Ashes (Fig. 19) and the rain-water on May 16 (Fig. 30), obtained by the similar procedure, one can find at once that the peaks in the curves corresponding to each species differ considerably, especially for anions and alkali earths. From these results it seems probable that the radioactivities in the rain-water have been caused by the nuclear detonation which might take place a few weeks before. However, this estimation should be ascertained by the absolute measurement of activities for each nuclides, which, to our regret, could not be carried out owing to insufficient data concerning several factors necessary for the absolute measurement.

VI. ACKNOWLEDGEMENT

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